Nodal gap structure of the heavy-fermion superconductor URu_2Si_2 revealed by field-angle-dependent specific-heat measurements

2017 J. Phys.: Conf. Ser. 807 052001

(http://iopscience.iop.org/1742-6596/807/5/052001)

Nodal gap structure of the heavy-fermion superconductor URu_2Si_2 revealed by field-angle-dependent specific-heat measurements

Shunichiro Kittaka[1], Yusei Shimizu[1], Toshiro Sakakibara[1], Yoshinori Haga[2], Etsuji Yamamoto[2], Yoshichika Ōnuki[2,3], Yasumasa Tsutsumi[4,5], Takuya Nomoto[6], Hiroaki Ikeda[7], and Kazushige Machida[7]

[1]Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan
[2]Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
[3]Department of Physics, University of the Ryukyus, Nishihara, Okinawa 903-0213, Japan
[4]Department of Basic Science, University of Tokyo, Tokyo 153-8902, Japan
[5]Condensed Matter Theory Laboratory, RIKEN, Wako, Saitama 351-0198, Japan
[6]Department of Physics, Kyoto University, Kyoto 606-8502, Japan
[7]Department of Physics, Ritsumeikan University, Kusatsu, Shiga 525-8577, Japan

E-mail: `kittaka@issp.u-tokyo.ac.jp`

Abstract. URu_2Si_2 is a promising candidate for a chiral d-wave superconductor whose gap is composed of a horizontal line node at equator and point nodes at the north and south poles. However, previous measurements of the specific heat and the thermal conductivity did not detect quasiparticle excitations from a horizontal line node in heavy-mass bands. Here, we have provided strong evidence for the presence of a horizontal line node from field-angle-dependent specific-heat measurements by using a high-quality single crystal of URu_2Si_2. We observed the $\sqrt{H}$ behavior of the specific heat at low fields which does not depend on inplane field angle ϕ but shows a shoulder-like anomaly in the polar-angle θ dependence around $\theta \sim 45°$. This feature is more clearly seen at lower temperatures, suggesting the detection of low-energy quasiparticle excitations reflecting the gap structure. From theoretical analyses based on microscopic calculations, we have demonstrated that this anomaly can be explained by the existence of a horizontal line node at $k_z = 0$. Thus, the gap structure of URu_2Si_2 matches well with the E_g chiral $k_z(k_x + ik_y)$ symmetry.

1. Introduction

URu_2Si_2 exhibits unconventional superconductivity below $T_c = 1.4$ K in the mysterious "hidden order" phase. Reflecting the likely exotic pairing mechanism, its superconducting gap appears to be anisotropic; the presence of nodes has been indicated from power-law temperature dependences of thermodynamic quantities such as the specific heat and the nuclear relaxation rate [1, 2, 3]. In addition, invariance of the specific heat [4] as well as the thermal conductivity [5] under a magnetic field rotated within the ab plane supports that the superconducting gap of URu_2Si_2 is rotationally symmetric around the c axis. From these results, E_g chiral d-wave superconductivity described by the gap symmetry of $k_z(k_x + ik_y)$ has been expected to be realized in URu_2Si_2. Indeed, broken time reversal symmetry in the superconducting state has been suggested from recent polar Kerr-effect measurements [6].

In the anticipated chiral d-wave state, the superconducting gap has point nodes at the north and south poles and a horizontal line node at $k_z = 0$. However, the results of specific-heat and thermal-conductivity measurements reported previously [3, 4, 5] cannot detect a horizontal line node in heavy-mass bands. At the time, it had been considered that heavy-mass Fermi surfaces are absent at $k_z = 0$ in the hidden order phase, resulting in disappearance of nodal quasiparticle excitations from the horizontal line node. Nevertheless, it has recently been clarified that heavy-mass bands similar to those in the antiferromagnetic phase [7, 8] exist at $k_z = 0$ also in the hidden order phase [9]. Therefore, the absence of quasiparticle excitations from the horizontal line node is incompatible with the chiral d-wave state.

In order to settle this controversy on the gap structure of URu_2Si_2, we have performed field-angle-dependent specific-heat measurements at low temperatures by using a high-quality single crystal [10]. In this contribution, we report the field-angle ϕ and θ dependence of the $\sqrt{H}$ behavior in the specific heat, evidencing the presence of a horizontal line node in heavy-mass bands at $k_z = 0$. These results ensure that the gap symmetry of URu_2Si_2 is indeed of the $k_z(k_x + ik_y)$ type.

2. Experimental

Single crystals of URu_2Si_2 were grown by the Czochralski pulling method in a tetra-arc furnace [11]. A high-quality single crystal (10.6 mg weight), the same sample in Ref. [10], was used in the present study. The specific heat C was measured by the standard quasi-adiabatic heat-pulse method or the relaxation method. The sample was cooled down to 0.1 K in a dilution refrigerator. Magnetic field was generated by using a vector magnet, up to 5 T in the x direction and 3 T in the z direction. The rotation of a refrigerator by using a stepper motor enables to control the field orientation three dimensionally with a high accuracy of $\sim 0.01°$. The field orientation is described by the azimuthal angle ϕ from the a axis and polar angle θ from the c axis.

3. Results and Discussion

Solid circles in Fig. 1 represent temperature dependence of the specific heat divided by temperature, C/T, of the present sample. For comparison, the C/T data taken from previous reports [4, 11, 12] are also plotted in the same figure. It is evident that the present sample is of the highest quality among them, as demonstrated by a sharp superconducting transition, high T_c, and low value of the residual C/T at low temperatures. Below 0.2 K, C/T shows anomalous increase on cooling. This upturn is too large to be attributed to a nuclear specific-heat contribution. Because the upturn is more prominent in a lower-quality sample [11], it might be caused by inclusion of some impurities.

Figure 2 plots the C/T data at 0.2 K as a function of $\sqrt{H}$ for $H \parallel a$ and $H \parallel c$. In both field directions, $C(H)$ is proportional to $\sqrt{H}$ at low fields, as represented by dashed lines. In the vortex state, energy spectra of quasiparticles are shifted due to the Doppler effect by $\delta E = m_e \boldsymbol{v}_F \cdot \boldsymbol{v}_s$, where m_e is quasiparticle mass, $\boldsymbol{v}_F$ is the Fermi velocity, and $\boldsymbol{v}_s$ is the velocity of supercurrent flowing around vortices. Around nodes, even a small shift of the spectra causes a finite zero-energy density of states (ZEDOS) which increases proportionally with $\sqrt{H}$ [13], in sharp contrast to the H-linear behavior expected Therefore, the observed $\sqrt{H}$ behavior in the low-temperature specific heat, which is a powerful probe to detect ZEDOS, demonstrates the presence of nodes in the superconducting gap.

In general, the field-orientation dependence of this $\sqrt{H}$ behavior reflects the location of gap nodes. When a magnetic field is pointing to a nodal direction, quasiparticle excitations at this node are strongly suppressed because of $\delta E = 0$, i.e., $\boldsymbol{v}_F \perp \boldsymbol{v}_s$. Therefore, the fact that the $\sqrt{H}$

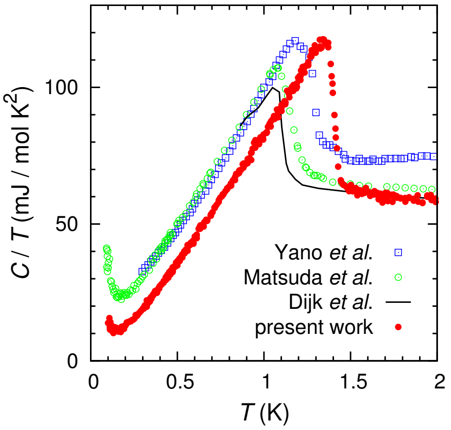

Figure 1. Temperature dependence of the zero-field C/T of the present sample (solid circles), compared with those of other samples in previous reports [4, 11, 12].

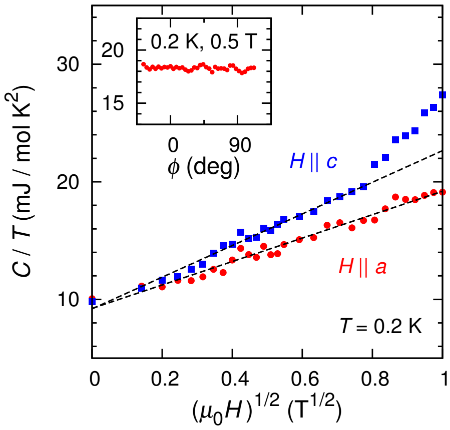

Figure 2. C/T as a function of $\sqrt{H}$ measured at 0.2 K for $H \parallel a$ and $H \parallel c$. Solid lines are fitting results for the low-field data by using a function $a\sqrt{H} + b$. Inset represents the azimuthal field-angle ϕ dependence of C/T at 0.2 K in a magnetic field of 0.5 T rotated within the ab plane.

behavior of $C(H)$ is observed in any field direction excludes a possibility of the gap structure having point nodes at the north and south poles alone [4]. Under a rotating magnetic field across nodal directions, this anisotropic Doppler effect causes an oscillation of the ZEDOS which shows a local minimum for a nodal direction. The inset of Fig. 2 presents the C/T data measured under a rotating field of 0.5 T within the ab plane at 0.2 K, where ϕ denotes the azimuthal field angle measured from the a axis. Any oscillation has not been observed, suggesting that vertical line nodes are not present in the gap.

The absence of vertical line nodes motivates us to search for a horizontal line node. In order to examine its presence and location, we measured the field-angle θ dependence of the specific heat, where θ is a polar angle between the magnetic field and the c axis. Figures 3 and 4 show $C(\theta)/T$ taken at 0.2 and 0.34 K, respectively. As shown in the inset of Fig. 4, $C(\theta)$ measured in the normal state at 1.8 K and 0.2 T is invariant with changing θ. At a low temperature of 0.2 K, a shoulder-like anomaly has been found around $\theta \sim 45^\circ$ in $C(\theta)$ measured in a field of 0.2 T. This anomaly moves to a larger θ with increasing field and becomes obscure by increasing temperature. The latter feature implies that this anomaly indeed comes from low-energy quasiparticle excitations around nodes.

To clarify the origin of the shoulder-like anomaly in $C(\theta)$, we have performed microscopic calculations on the basis of the quasiclassical Eilenberger theory assuming a chiral d-wave gap of the $k_z(k_x + ik_y)$ type and a spherical Fermi surface [14]. In these calculations, the Pauli-paramagnetic effect and the anisotropy of the Fermi velocity have not been considered. Figures 5(a) and 6(a) show the polar-angle θ dependence of the ZEDOS, $N(E = 0)$, at $B = 0.03$ and 0.1, respectively. Here, B is scaled by the Eilenberger unit [15]. Note that Fig. 5(a) reproduces the shoulder-like anomaly at $\theta \sim 45^\circ$. With increasing field, the shoulder-like anomaly moves to higher θ and a clear dip appears at $\theta = 0^\circ$ due to reversal of the $C(\theta)$ anisotropy originating from the slight anisotropy of H_{c2} ($H_{c2}^{\parallel x} \gtrsim H_{c2}^{\parallel z}$) reflecting the gap structure. In the experiment, $C(\theta)$ of URu_2Si_2 does not show the dip at $\theta = 0^\circ$ because the strong Pauli-

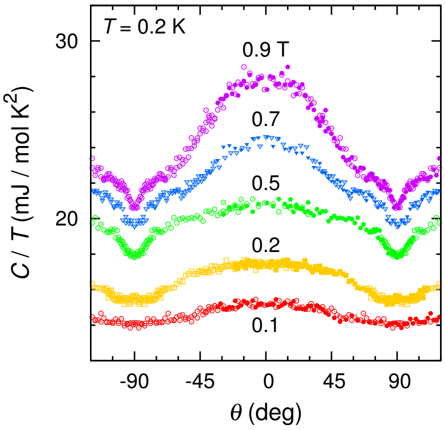

Figure 3. Polar field-angle θ dependence of C/T at 0.2 K in rotating fields within the ac plane. Numbers labelling curves represent the applied magnetic field in tesla.

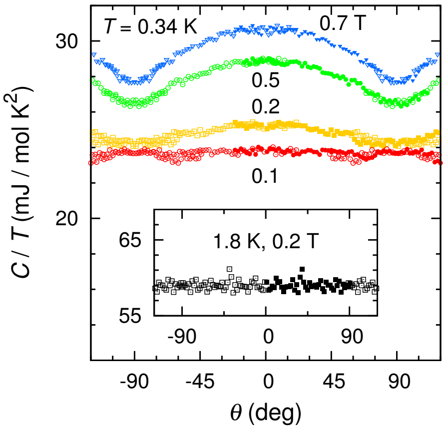

Figure 4. The $C(\theta)/T$ data at 0.34 K in several fields. Inset shows the normal-state $C(\theta)/T$ measured at 1.8 K in 0.2 T.

paramagnetic effect yields a peak at $\theta = 0°$.

In Figs. 5(b) and 6(b), we show the $\boldsymbol{k}$-resolved density of states, $N_{\boldsymbol{k}}(0)$, mapped on the spherical Fermi surface at several θ for $B = 0.03$ and 0.1 rotated within the xz plane, respectively. Here, the integration of $N_{\boldsymbol{k}}(0)$ over the Fermi surface corresponds to $N(E = 0)$. Most of quasiparticle excitations occur around a line node; contribution from point nodes to $N(E = 0)$ is relatively small. Although $N_{\boldsymbol{k}}(0)$ is enhanced in the overall Fermi surface by increasing magnetic field, qualitatively similar features have been seen in both fields; $N_{\boldsymbol{k}}(0)$ is strongly suppressed at the k_x direction when θ becomes larger than 45°, i.e., for $\theta = 75°$ and 90°. This is due to the suppression of the Doppler effect there and yields the shoulder-like anomaly in $C(\theta)$. Because the low-temperature specific heat is dominated by heavy quasiparticles, the shoulder-like anomaly in $C(\theta)$ evidences the presence of a horizontal line node at $k_z = 0$ in heavy-mass bands. This fact further supports that the gap symmetry of URu_2Si_2 belongs to the chiral E_g symmetry of the $k_z(k_x + ik_y)$ type [16].

4. Summary

We report the results of field-angle-dependent specific-heat measurements by using a high quality single crystal of URu_2Si_2. It was found that the specific heat shows the $\sqrt{H}$ behavior in any field direction and the shoulder-like anomaly in its polar-angle θ dependence which is smeared out with increasing temperature. From theoretical analyses based on the microscopic theory, we have demonstrated that these features are strong evidence for the presence of a horizontal line node in the heavy-mass bands. Thus, the present study settles the remaining controversy in URu_2Si_2 and helps establishing that URu_2Si_2 is a chiral d-wave superconductor of the $k_z(k_x + ik_y)$ type.

Acknowledgement

We thank Y. Yanase for useful discussions. A part of the numerical calculations was performed by using the HOKUSAI GreatWave supercomputer system in RIKEN. This work was supported by a Grants-in-Aid for Scientific Research on Innovative Areas "J-Physics" (15H05883) from MEXT, and KAKENHI (15K05158, 15H03682, 25103716, 26400360, 15H05745, 15K17715, 15J05698) from JSPS.

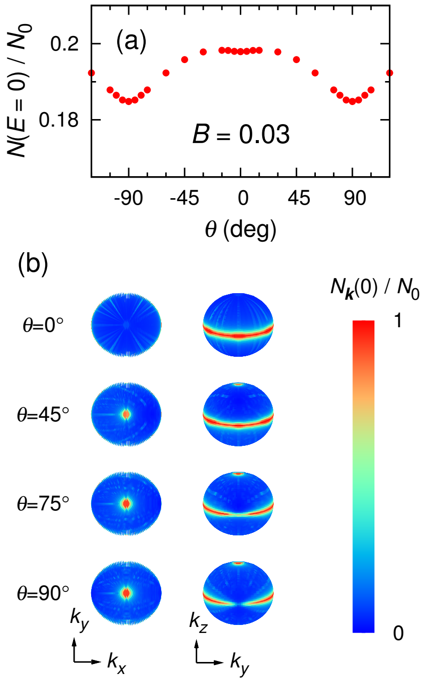

Figure 5. (a) Polar field-angle θ dependence of $N(E=0)$ at $B=0.03$ scaled by the normal-state density of states, N_0, obtained by microscopic calculations with the assumption of the gap symmetry of $k_z(k_x+ik_y)$. (b) Angle-resolved density of states $N_{\boldsymbol{k}}(0)$ at $B=0.03$ for selected θ, mapped on the spherical Fermi surface. Here, the magnetic field is rotated within the xz plane.

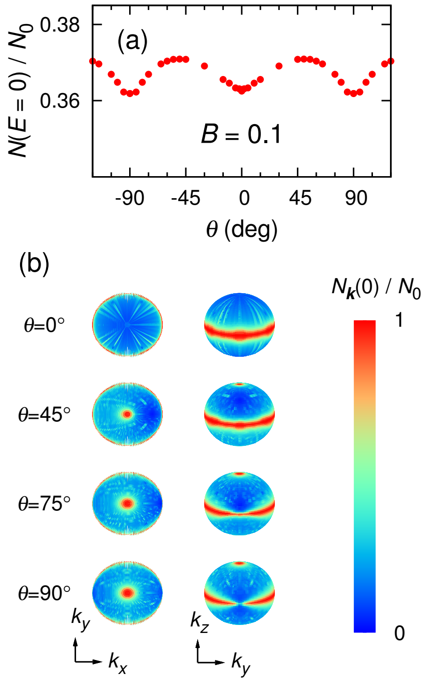

Figure 6. (a) θ dependence of $N(E=0)/N_0$ at $B=0.1$. (b) $N_{\boldsymbol{k}}(0)$ for selected θ in a rotating magnetic field of $B=0.1$, mapped on the spherical Fermi surface.

References

[1] Fisher R A, Kim S, Wu Y, Phillips N E, Mcelfresh M, Torikachili M S and Maple M B 1990 *Physica B* **163** 419
[2] Matsuda K, Kohori Y and Kohara T 1996 *J. Phys. Soc. Jpn.* **65** 679
[3] Kasahara Y, Iwasawa T, Shishido H, Shibauchi T, Behnia K, Haga Y, Matsuda T D, Onuki Y, Sigrist M and Matsuda Y 2007 *Phys. Rev. Lett.* **99** 116402
[4] Yano K, Sakakibara T, Tayama T, Yokoyama M, Amitsuka H, Homma Y, Miranović P, Ichioka M, Tsutsumi Y and Machida K 2008 *Phys. Rev. Lett.* **100** 017004
[5] Kasahara Y, Shishido H, Shibauchi T, Haga Y, Matsuda T D, Onuki Y and Matsuda Y 2009 *New J. Phys.* **11** 055061
[6] Schemm E R, Baumbach R E, Tobash P H, Ronning F, Bauer E D and Kapitulnik A 2015 *Phys. Rev. B* **91** 140506(R)
[7] Ohkuni H, Inada Y, Tokiwa Y, Sakurai K, Settai R, Honma T, Haga Y, Yamamoto E, Yamagami H, Takahashi S and Yanagisawa T 1999 *Phil. Mag. B* **79** 1045
[8] Ikeda H, Suzuki M T, Arita R, Takimoto T, Shibauchi T and Matsuda Y 2012 *Nat. Phys.* **8** 528
[9] Hassinger E, Knebel G, Matsuda T D, Aoki D, Taufour V and Flouquet J 2010 *Phys. Rev. Lett.* **105** 216409
[10] Kittaka S, Shimizu Y, Sakakibara T, Haga Y, Yamamoto E, Onuki Y, Tsutsumi Y, Nomoto T, Ikeda H and Machida K 2016 *J. Phys. Soc. Jpn.* **85** 033704

[11] Matsuda T D, Hassinger E, Aoki D, Taufour V, Knebel G, Tateiwa N, Yamamoto E, Haga Y, Ōnuki Y, Fisk Z and Flouquet J 2011 *J. Phys. Soc. Jpn.* **80** 114710
[12] van Dijk N H, Bourdarot F, Klaasse J C P, Hagmusa I H, Brück E and Menovsky A A 1997 *Phys. Rev. B* **56** 14493
[13] Volovik G E 1993 *JETP Lett.* **58** 469
[14] Tsutsumi Y, Nomoto T, Ikeda H, and Machida K, arXiv:1604.02806.
[15] Miranović P, Ichioka M, Machida K and Nakai N 2005 *J. Phys.: Condens. Matter* **17** 7971
[16] Sigrist M and Ueda K 1991 *Rev. Mod. Phys.* **63** 239